

Excitable Dynamics in High-Lewis Number Premixed Gas Combustion at Normal and Microgravity

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Abstract

Freely-propagating, premixed, gas flames in high-Lewis (Le) number, quiescent mixtures are studied experimentally in tubes of various diameter at normal (1g) and microgravity (μg). A premixture of lean butane and oxygen diluted with helium, argon, neon, nitrogen or a mixture of multiple diluents is examined such that the thermal diffusivity of the mixture (and to a lesser extent, the mass diffusivity of the rate-limiting component) is systematically varied. In effect, different diluents allow variation of the Le without changing the chemistry. The flames are recorded with high speed cinematography and their stability is visually assessed.

Different modes of propagation were observed depending on the diameter of the tubes (different conductive heat loss), the composition of the mixture and the g-level. At 1g, four modes of propagation were observed in small and intermediate diameter tubes (large conductive heat loss): (1) steadily propagating flames, (2) radial and longitudinal pulsating flames, (3) "wavering" flames and (4) rotating spiral flames. As the diameter of the tube increases, the radial modes become more pronounced while the longitudinal modes systematically disappear. Also, multiple, simultaneous, spatially-separated "pacemaker" sites are observed in intermediate and large diameter tubes. Each site starts as a small region of high luminosity and develops into a flamelet which assumes the form of one of the forementioned modes. These flamelets eventually interact, annihilate each other in their regions of intersection and merge at their newly created free-ends. For very large tubes, radially-propagating wave-trains (believed to be "trigger waves") are observed. These are analogous to the radial pulsations observed in the smaller diameter tubes. At μg , three modes of propagation have been observed: (1) steadily propagating flames, (2) radial and longitudinal pulsating flames and (3) multi-armed, rotating flames.

Since the pulsating mode exists at μg and 1g, buoyant flicker is not the mechanism which drives the pulsations. Moreover, all of the instabilities at 1g and μg have characteristic frequencies on the $O(100\text{Hz})$. This value is lower than the fundamental, longitudinal acoustic frequencies of the tubes which suggests that the instabilities are not acoustically driven.

The patterns formed by this reaction bear remarkable similarities with the patterns formed in most excitable media when the behavior of the system is driven by couplings between chemical reaction and diffusion (e.g., Belousov-Zhabotinsky reaction, patterns in slime molds, spiral waves in the retina of a birds eye). While it is recognized that the chemical mechanism associated with this premixed gas reaction is exponentially sensitive to temperature and undoubtedly different from those which govern previously observed excitable media (most are isothermal, or weakly exothermic, liquid phase reactions), similar spatial and temporal patterns should not come as a complete surprise considering heat and mass diffusion are self-similar.

It is concluded that this premixed gas system is a definitive example of a diffusive-thermal, gas-phase oscillator based on these experimental results and their favorable comparison with theory.

1. Introduction

To verify the existence of the high- Le number diffusive-thermal instability in premixed gas combustion, μg tests were initially performed using a mixture of butane and oxygen diluted with helium ($Le=3$) in a flame tube opened at the ignition-end and closed at the other end. Based on a scale analysis, the frequency of the oscillation (ω) is expected to scale with the square of the laminar burning velocity divided by the thermal diffusivity of the mixture and the amplitude of the oscillation (δ) is expected to scale with the thermal diffusivity divided by the burning velocity. At μg , the laminar burning velocity in near-limit mixtures is not affected by buoyant convection and thus may be lower than the burning velocity at 1g. Hence, by reducing the flame speed, ω should decrease and δ should increase, making it easier to see. This idea, originally proposed by Ronney [1], seemed plausible considering the only experimental evidence to support the existence of the high- Le number, diffusive-thermal instability [2] was complicated by complexities of heat loss to the burner and sensitivity to the local hydrodynamic field. By considering freely-propagating flames in quiescent mixtures, rather than burner-stabilized flames, the heat loss and hydrodynamic problems are eliminated. Also, at μg , buoyant convection is eliminated as a potential source of any observed instability. This was the starting point approximately two years ago.

During the past two years, we learned several new things about the high- Le number diffusive-thermal instability:

1. The instability exists at 1g and μg .

2. The instability manifests itself in several different forms depending on the composition of the mixture (hence, Le), the conductive heat loss to the walls, and the g -level.
3. A critical Le_c (>1) exists. Below Le_c (but still >1), the flames propagate in a stable, uniform manner. Above Le_c , the flames are unstable.

These findings are described in this paper. The μg experimental apparatus and results is presented in Section 2 followed by the $1g$ tests on downwardly propagating flames in Section 3. A brief summary and description of work-in-progress is presented in Section 4.

2. Microgravity Apparatus and Results

Freely-propagating flames are studied experimentally at μg in the 2.2 sec drop tower at the NASA Lewis Research Center [3]. The tests are performed in a 14.3cm i.d., polycarbonate flame tube, 40.0cm in length. The tube is permanently closed at one end and initially sealed with a mylar membrane at the opposite end. A hot-wire/ nitrocellulose igniter is situated on the nonreactive side of the membrane. The resistively heated wire ignites the nitrocellulose releasing approximately 40.0J. This is sufficient to completely consume the membrane, thereby open the ignition-end to ambient conditions, and ignite the mixture. The flame shape and its evolution are recorded at 200frames/sec with a high-speed Millikan 16mm movie camera. Although higher framing rates are desirable, the visible light from the flame is insufficient to leave a trace on the film at rates greater than 250 frames/sec, even with the most sensitive film. A schematic of the drop rig is shown in Fig. 1.

The tube is filled by the partial pressure method to local atmospheric pressure, 740-750 Torr, with a mixture of C_4H_{10} - O_2 -He. A fan is installed on the closed end of the tube to mix the reactants prior to ignition. The fan is turned on during the fill procedure and it is turned off approximately five minutes prior to the drop. This is about five times longer than a characteristic viscous dissipation time (53 sec) based on the ratio of the square of the radius of the tube to the kinematic viscosity of helium at room temperature.

Careful scrutiny of the film suggests that a multi-armed, rotating wave and a radial and longitudinally pulsating flame exist as the flammability limit is approached. The characteristic temporal frequencies of both the pulsating and traveling waves are on the order of 70-100 cycles/s.

Despite the limited camera resolution, three distinct modes of propagation can be identified according to the volumetric fuel concentration in the mixture,

- (1) *Stable* propagating flames occur in mixtures containing 1.30% (or more) C_4H_{10} ,
- (2) *Rotating waves* occur in mixtures containing 1.21-1.22% C_4H_{10} , and
- (3) *Combined radial and longitudinal pulsations* occur in mixtures containing 1.21-1.25% C_4H_{10} .

Figure 2 shows an axial view of a radial and longitudinally pulsating flame. Each pulsation cycle starts with a bright, centrally-located, "pacemaker" region. This bright region radially expands and decreases in luminosity as the flame propagates axially into the reactants. As the flame nears its minimum intensity and maximum surface area (when it approaches the wall), another bright region appears at (or near) the center and the cycle repeats. Although it is not clear whether these bright zones are the flame or are traveling on the surface of a much less visible flame, the latter is presently speculated to be the case since it would otherwise be difficult to justify why the radial propagation speed (as high as 2.0m/s) is roughly ten times the axial flame propagation speed. One possible argument, proposed by Art Winfree, suggests that the phase of the oscillation may vary from one part of the flame to another where the "observed" speed is proportional to the inverse of the phase gradient. This would allow for infinitely large radial speeds.

The rotating wave (Fig. 3) observed at μg consists of six periodic, azimuthally-distributed bright and dark zones which rigidly rotate around the axis of the tube, tracing out a helical trajectory as the flame axially propagates into the reactants. While this mode appears to be unique to μg , additional tests are planned to determine whether a correlation exists between this multi-armed structure and a multi-armed, rotating spiral wave observed at $1g$ (discussed in Section 3).

3. Normal-Gravity Apparatus and Results

Three different internal diameter (i.d.) Plexiglas tubes (5.1, 10.2 or 14.3cm), approximately 1.2m in length, are used to study downwardly propagating flames at $1g$. The bottom of the tubes is permanently closed with a Plexiglas window while the other end is initially closed with a plate connected to a solenoid. When the solenoid is activated, the plate is lifted vertically off of an o-ring seal. Similar to the μg tests, a hot wire/ trinitrocellulose igniter is situated at the open-end (top).†

The tube is filled by the partial pressure method or mass flow rate with a mixture of C_4H_{10} , O_2 and an inert (or inert mixture). A fan is attached to the removable end plate and used to ensure homogeneous mixing of the reactants. It is turned off approximately five minutes after the last constituent is added at which time the mixture is allowed ten minutes to quiesce prior to ignition; this waiting period is at least ten times the viscous dissipation time (53 sec).

† Additional tests were performed using the same tubes with a membrane and igniter at the open-end (similar to the μg tests) to eliminate the possibility of any convective flow induced by the movable end plate.

High-speed video is obtained using two Kodak EktaPro intensified video cameras each recording the flame at 1000 frames/sec. One camera is axially positioned in the middle of the tube (field of view is 20cm) while a second camera obtains a radial view from a mirror positioned at a 45° angle with respect to the base (closed-end) of the tube. The video outputs are recorded using a high-speed tape drive and then downloaded at 30 frames/sec to VHS video.

A summary of the "instabilities" is given for each tube diameter. The word "instabilities" is in quotes because it can be a misnomer; these modes of propagation are quasi-stable for most experimental conditions, except for exceptionally near-limit flames which spontaneously transition between modes.

Also, note that all of the observed "instabilities" exist within approximately 0.2 molar percent (or less) C_4H_{10} from the lean flammability limit (LFL) where the LFL's are determined experimentally to be 1.52, 1.44 and 1.29 percent C_4H_{10} in a 21% O_2 -He environment for the 5.1, 10.2 or 14.3cm i.d. tubes, respectively. Conductive heat loss apparently governs the limit in the small tubes whereas buoyant convection and radiative losses govern the limit in the larger diameter tubes.

5.1cm i.d. tube: In the smallest tube tested, four modes of propagation (Fig. 5) are observed:

- (a) stable, uniformly-propagating, axisymmetric flames in mixtures containing 1.68% C_4H_{10} (or more),
- (b) longitudinal, axisymmetric pulsations ($\omega \sim 170$ -200Hz) in mixtures containing 1.60-1.67% C_4H_{10} , and
- (c) tristable modes of propagation in mixtures containing 1.52-1.59% C_4H_{10} . The flame alternates between three modes: (1) a longitudinal pulsation (Fig. 5b), (2) a "wavering" mode, characterized by a bright spot on the flame swaying from one side of the flame to the other, tracing out a nonaxisymmetric, zig-zag path (Fig. 5c), or (3) a rotating spiral wave (Fig. 5d). While the longitudinal oscillations are easy to visualize, the "wavering" mode and spiral wave deserve some attention. By propagating in a zig-zag or spiral path, the flame can gain a foothold in the reactants without having to propagate in discrete, longitudinal steps. In this way, the flame can propagate in a spatially continuous fashion. Interestingly, many crystals grow in an analogous way [4]. The molecules escape from the solid into the surrounding vapor at a rate dependent on the surface temperature and the molecules condense on the surface at a rate proportional to the vapor concentration. At very slight supersaturations (as low as 1%), crystals grow in a spiral ramp or a series of interlaced, zig-zagging concentric rings, due to pure diffusion. Rather than counterdiffusion of a single species, counterdiffusion of heat and mass apparently induce similar spatial patterns in our premixed gas flame.

10.3cm i.d. tube Four modes are also observed in a tube with twice the i.d. (Fig. 6):

- (a) stable, uniformly propagating, axisymmetric flames in mixtures containing 1.56% C_4H_{10} (or more),
- (b) combined radial and longitudinal pulsations in mixtures containing 1.48±0.01 to 1.54% C_4H_{10} (while the amplitude of the longitudinal pulsation is measurably smaller than in the 5.1cm i.d. tube),
- (c) trinary mode of propagation in mixtures containing 1.45 to 1.48±0.01% C_4H_{10} . Similar to the flames in the 5.1cm i.d. tube, the flame alternates between three modes: (1) a combined radial and longitudinal pulsation, (2) a "wavering" mode, or (3) a rotating spiral wave.

14.3cm i.d. tube Similar modes are observed in a larger diameter tube (Fig. 7a,b,c). Once again,

- (a) stable, uniformly propagating, axisymmetric flames occur in mixtures containing greater than 1.50% C_4H_{10} (or more),
- (b) radial pulsations occur in mixtures containing 1.35-1.50% C_4H_{10} , and
- (c) bistable modes of propagation occur in mixtures containing 1.29-1.35% C_4H_{10} . The flame alternates between a rotating spiral wave and a radial pulsation.

In addition, multiple, simultaneous pacemaker sites are observed at the same cross-section of the tube for these very near-limit mixtures. Each "site" develops into a radial pulsation or a rotating spiral wave (CW or CCW). Since the flames compete for the same reactants, they inevitably coalesce, annihilate one another in their regions of intersection and either merge at their free-ends or separate into flamelets which ultimately meet again. In Fig. 7d, the initiation and development of two counter-propagating radial pulsations are shown. In Fig. 7e, two co-rotating spiral waves have developed separately and then joined at their pivoting endpoints to form a two-armed rotating spiral wave.

30.5cm x 30.5cm x 153cm square duct: A few preliminary tests have been performed in a large square duct, open at the top and closed at the bottom. Mixtures containing 1.32 to 1.47% C_4H_{10} were tested, where the LFL has been determined to be 1.32% C_4H_{10} in a 21% O_2 -He environment (compare with the value of 1.29% in the 14.3 cm i.d. tube). For this range of experimental parameters, multiple, radial propagating wave-trains appear and propagate outward in a very systematic fashion. In the smaller tubes, these "wave-trains" appear as radial pulsations since the characteristic spacing between waves is on the order of the tube diameters. In other words, the radial propagating flames previously observed quench at the tube walls before another radial (concentric ring) wave develops.

These wave-trains are believed to be "trigger waves"--waves of chemical activity whose existence depends on the interaction of reaction and diffusion. They are characterized by the fact that they can be

blocked by impermeable barriers. Analogous waves have been observed in the Belousov-Zhabotinsky [5] reaction and should not be confused with kinematic waves (phase or frequency gradient waves) which are independent of diffusion [6]. Experimental evidence which supports this contention will be presented under separate cover.

4. Discussion and Work-in-Progress

To summarize the results so far, freely-propagating flames in C_4H_{10} - O_2 mixtures diluted with pure helium exhibit several different modes of propagation near the LFL. These modes have been classified according to the volumetric percent fuel in the mixture (i.e., equivalence ratio) from which the Le can be determined. Also, above a critical equivalence ratio, the flames propagate in a steady manner and do not visually display any evidence of "instability."

Preliminary tests have also been performed using four component mixtures where the fourth component is a second "inert" gas (Ne, Ar, N_2) such that the Le of the bulk mixture is systematically varied without varying the equivalence ratio. Experimentally, the percent He in the earlier mixtures is reduced and replaced with an equivalent percent of an additional inert. These results indicate that flames which are "unstable" (do not propagate in a steady manner) in a pure He-diluted mixture stabilize above a critical $[He]/[diluent]$ ratio. This suggests that the Le of the bulk mixture must be sufficiently greater than one for onset of "instability." Stability diagrams (Le vs. conductive heat loss) are currently being constructed where the values of the mixture Le 's are evaluated using CHEMKIN thermodynamic transport data as input into the Sandia multicomponent transport code.

As mentioned before, the spatio-temporal patterns formed by this premixed gas flame rival those observed in other excitable media. Both systems display rotating spiral waves and concentric ring waves. Initially, these ring waves in our premixed system appeared to be pulsations since the wavelength (spacing between rings) is on the order of the tube diameter. In addition, this premixed gas reaction displays simultaneous, multiple "pacemaker" sites which initially propagate as though they were independent entities. They eventually collide and quench in their regions of intersection, due to local depletion of the rate-limiting component (C_4H_{10}), and then continue to develop in the form of a combined entity or break-up into distorted shapes. This is representative of chemical turbulence.

Perhaps, it is also worth noting that analogous modes of propagation have been observed in solid flames [7] which also have very large Le 's due to their very small diffusivities. Those who have studied these phenomena have seen steady flames, auto-oscillations, spinning spiral waves, and "chaotic" modes. Interestingly, the "chaotic" modes appear when points in the combustion wave move around independently of one another. These phenomena may be similar to the thermally-driven instabilities that are observed in our premixed gas flames, where the "chaotic" mode seen in the solid flames may correspond to the existence of multiple, simultaneous pacemaker sites which subsequently perform their own dynamics. Unfortunately, one cannot easily see the face of the moving front in the solid phase.

Acknowledgments

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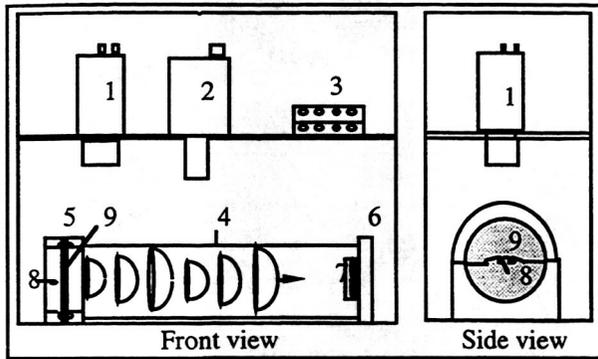


Fig. 1.- Microgravity drop apparatus (1) Standard (30 frame /sec) Video Camcorder, (2) Millikan High Speed 16mm Camera, (3) Electronics, (4) Flame Tube, (5) Ignition-End, (6) Closed End, (7) DC Fan, (8) Igniter, (9) Membrane

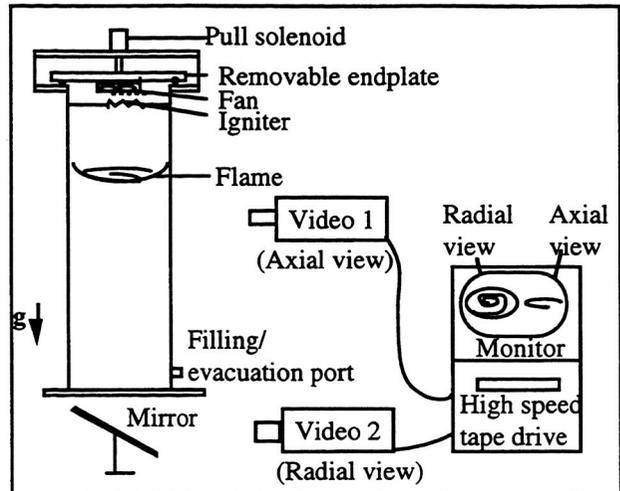


Fig. 4. - Normal-Gravity Experimental Apparatus

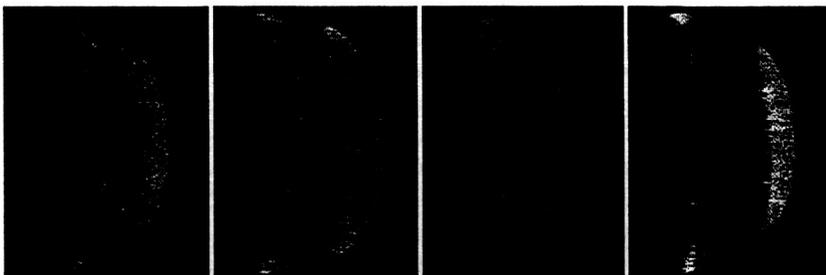


Fig. 2: Pulsating flame at μg in 1.21% C_4H_{10} -21.0% O_2 -He. The time between sequential frames is 1/200 sec.

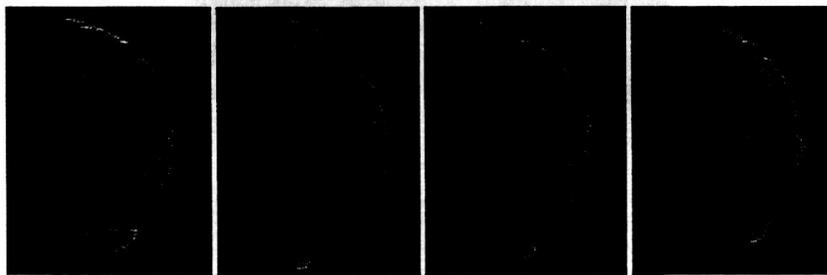


Fig. 3: Spinning propagation at μg in 1.21% C_4H_{10} -21.0% O_2 -He. The time between sequential frames is 1/200 sec.

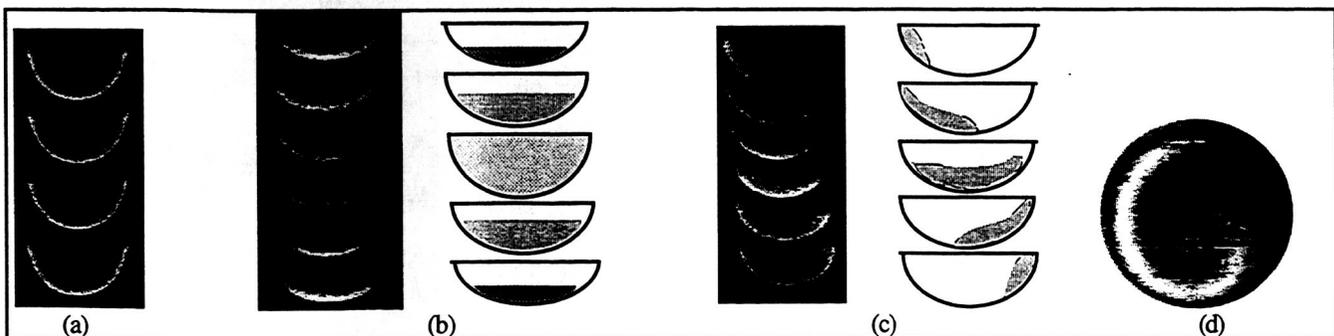


Fig. 5.- (a) stable flame in 1.69% C_4H_{10} -21% O_2 -He, (b) longitudinal oscillations in a mixture containing 1.63% C_4H_{10} -21% O_2 -He, (c) "wavering" flame in a mixture containing 1.53% C_4H_{10} -21% O_2 -He (axial view), (d) radial view of a rotating spiral wave in a mixture containing 1.53% C_4H_{10} -21% O_2 -He; the horizontal bands are due to a tracking problem with the high speed video. The time between sequential images is 1/1000s.

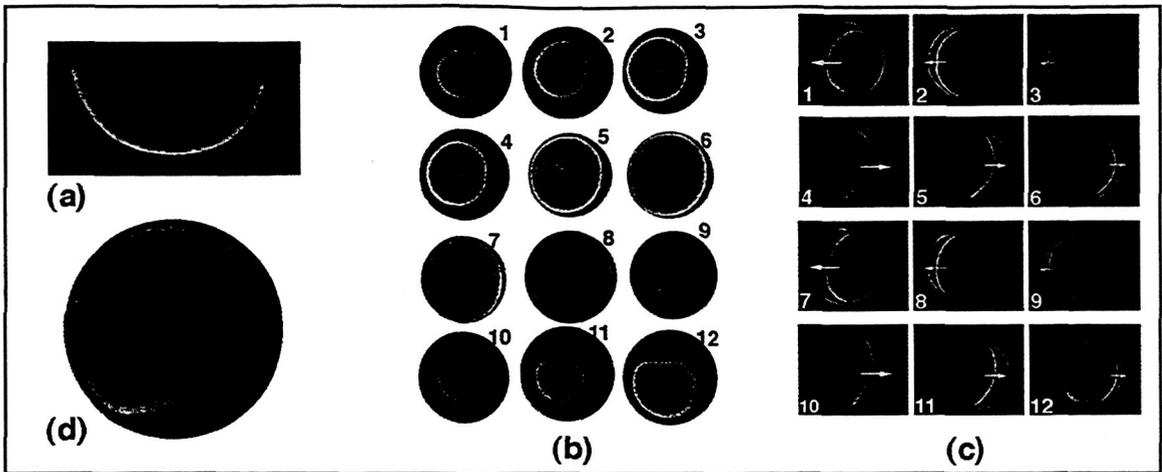


Fig. 6.- (a) stable flame in 1.56% C_4H_{10} -21% O_2 -He, (b) radial oscillations in 1.45% C_4H_{10} -21% O_2 -He (radial view), (c) "wavering" flames in 1.45% C_4H_{10} -21% O_2 -He (radial view); arrows indicate direction of propagation; axial view is similar to Fig. 5c, (d) radial view of spiral flame in 1.45% C_4H_{10} -21% O_2 -He. The time between sequential images is 1/1000s in (b) and 1/200s in (c).

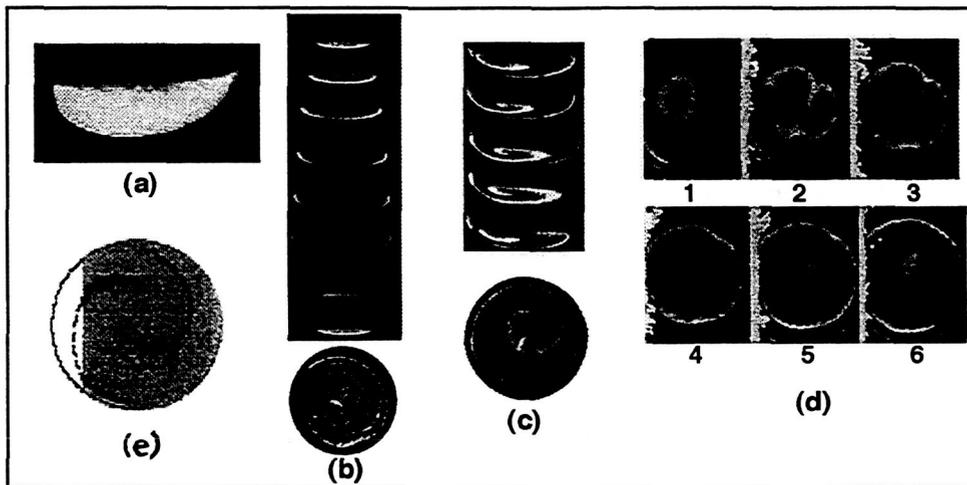


Fig. 7.- (a) stable flame in 1.50% C_4H_{10} -21% O_2 -He (axial view), (b) radial pulsations in mixture containing 1.35% C_4H_{10} -21% O_2 -He (axial and radial views), (c) spiral wave in a mixture containing 1.30% C_4H_{10} -21% O_2 -He (axial and radial views), (d) the intersection and evolution of two simultaneous, radial pulsations (radial view), (e) negative image a multi-armed rotating spiral wave (radial view). Time between sequential images is 1/1000s in (b and c) and 1/500s in (d).

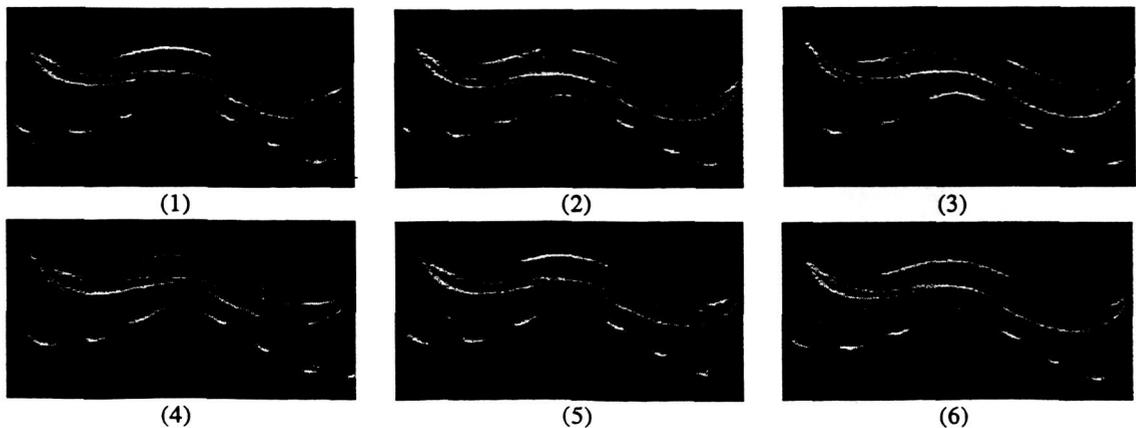


Fig.8.-Wave-trains (believed to be "trigger-waves") emanating from the center are observed in large diameter vessels. The mixture is 1.34% C_4H_{10} -21% O_2 -He. The time between sequential images is 1/1000s.